Phase Effects in Two-Photon Free-Free Transitions in a Bichromatic Field of Frequencies ω and 3ω

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Abstract. The effect of the relative phase between the components of a bichromatic field of frequencies ω and 3ω is discussed in the case of free-free transitions in laser-assisted electron-hydrogen scattering. For fast projectile and low field intensities, the role of target dressing is pointed out.

I. INTRODUCTION

High harmonic generation techniques have recently been used to provide intense sources of multichromatic coherent radiation (L'Huillier et al 1992). Atomic and molecular systems are submitted to such radiation fields in order to get new insights into the dynamics of laser assisted processes. Also, they may represent sensitive "tools" (Véniard et al 1996), which allow us to investigate a key parameter in high harmonic generation phenomenon: the phase difference between the harmonics.

Free-free transition in laser-assisted electron-atom scattering in a bichromatic field is a process in which the phase difference is a sensitive parameter. Theoretical investigations on this topic have recently been published. The early results were obtained for low frequencies, neglecting the dressing of the target (Varró and Ehlotzky 1993, 1993a, and Ehlotzky 1994). A major aspect investigated in these works was the influence of the relative phase between the components of the bichromatic field on the laser assisted signals. However, perturbative calculations for both monochromatic (Dubois et al 1986, Kracke et al 1994) and bichromatic fields (Cionga & Buică 1998) have shown that the dressing of the target by the radiation field plays an important role when the field frequency is no longer small. The effect of target dressing on free-free transitions in a bichromatic field of frequencies ω and 2ω was investigated in the domain of moderate field intensities for fast projectiles: the laser-atom

interaction was described by first order perturbation theory (Varró and Ehlotzky 1997) or by second order perturbation theory (Cionga and Zloh 1999).

The aim of our work is to study two-photon free-free transitions in electron-hydrogen scattering when the radiation field is the superposition of two components of frequencies ω and 3ω:

$$\vec{\mathcal{A}}(t) = \vec{\varepsilon}\mathcal{A}_0 \cos\omega t + \vec{\varepsilon}\,'\mathcal{A}'_0 \cos(3\omega t + \varphi) \,. \tag{1}$$

$\mathcal{A}_0$ is the amplitude of the vector potential describing the laser field of frequency ω, $\mathcal{A}'_0$ describes the second harmonic of frequency $\omega' = 3\omega$; $\vec{\varepsilon}$ and $\vec{\varepsilon}\,'$ are the corresponding polarization vectors. φ is the relative phase between the harmonic and the fundamental field and we focus our attention on a systematic investigation of its influence on laser assisted signals which correspond to the scattered electrons with the final energy

$$E_f = E_i \pm 2\omega, \tag{2}$$

where $E_{i(f)}$ is the initial (final) energy of the projectile. In the presence of the radiation field (1) the energy (2) is reached by two different quantum paths. For the sake of simplicity, only the case $E_f > E_i$ is schematically represented in Fig.1. Two identical photons, of frequency ω, are absorbed in the process associated to the path labeled by (a): the projectile gains an energy equal to 2ω, since the internal state of the atom is not modified due to the scattering. On the other path, (b), the high frequency photon is absorbed and the fundamental one is emitted, leading to the same final energy: $E_f = E_i + 2\omega$. The relative phase φ "modulates" the quantum interference between the two paths, as it will be discussed in this paper.

In the domain of high scattering energies and low field intensities, we use the third order perturbation theory (second order in the electric field and first order in the scattering potential) to evaluate the differential cross section of the scattered electrons. The calculations are described in the Section II; they are carried out taking into account *all* the involved Feynman diagrams for each of the paths (a) and (b). This represents an appropriate treatment of two-photon free-free transitions, including the modification of the target in the field in second order perturbation theory. The case we study here implies the quantum interference of two processes involving the same numbers of photons, two. *In the energy spectrum of the scattered electrons this corresponds to the second pair of sidebands ($N = \pm 2$).* It is the

simplest case of this type in which phase effects may be investigated taking consistently into account the dressing of the target. *Although it involves second order processes, this case is more suitable for a discussion of interferences and phase effects than the first pair of sidebands ($N = \pm 1$). In that case, the interferences would involve one- and three-photon processes.* Our numerical results, presented in Section III, are obtained for identical linear polarizations, in the geometry in which the polarization vector, $\vec{\varepsilon}$, is parallel to the initial momentum of the projectile. We consider fast projectiles, $E_i = 100$ eV, and the frequency $\omega = 1.17$ eV, corresponding to Nd:YAG laser. Phase effects are investigated in the domain of small scattering angles, pointing out the influence of target dressing on these effects.

II. BASIC EQUATIONS

The time evolution of the electron-hydrogen system in the presence of the electromagnetic field described by Eq.(1) is governed by the hamiltonian

$$\mathcal{H} = \frac{\vec{P}^2}{2} - \frac{1}{R} + \frac{\vec{p}^{\,2}}{2} + \frac{1}{|\vec{r} - \vec{R}|} - \frac{1}{r} + \frac{1}{c}\left[\vec{p} + \vec{P}\right] \cdot \vec{\mathcal{A}}(t) \equiv H_0 + V + W(t), \tag{3}$$

where $\vec{R}$, $\vec{P}$ are the position and momentum operator of the bound (atomic) electron and $\vec{r}$, $\vec{p}$ are the position and momentum operator of the free (projectile) electron. $V \equiv -r^{-1} + |\vec{r} - \vec{R}|^{-1}$ denotes the e-H interaction in the direct channel, when exchange effects are neglected. $W(t) \equiv c^{-1}\left[\vec{p} + \vec{P}\right] \cdot \vec{\mathcal{A}}(t)$ denotes the interaction of the charge particles with the field, treated in the velocity gauge, using the dipole approximation. The $\vec{\mathcal{A}}^2$-term was eliminated through a unitary transformation.

In the *first nonvanishing order* of the perturbation theory, the $S-$ matrix elements corresponding to two-photon processes are given in second order perturbation theory by

$$S^{(2)} = -\int_{-\infty}^{+\infty} dt_1 \int_{-\infty}^{t_1} dt_2 < \chi_f^- | \tilde{W}(t_1) \tilde{W}(t_2) | \chi_i^+ >, \tag{4}$$

where $\tilde{W}(t) = e^{iH_0 t} W(t) e^{-iH_0 t}$. In the previous equation $|\chi_i^+ >$ and $|\chi_f^- >$ describe the initial and final states of the colliding system (electron-atom)

$$|\chi_i^+ > \; = \; |\Psi_i > + G^+(\mathcal{E}_i) V |\Psi_i >, \tag{5}$$

$$|\chi_f^- > \; = \; |\Psi_f > + G^-(\mathcal{E}_f) V |\Psi_f >, \tag{6}$$

where

$$G^{\pm}(\mathcal{E}) = [\mathcal{E} - H_0 - V \pm i\delta]^{-1} \tag{7}$$

and δ a positive infinitesimal number. $|\Psi_{i,f}>$ are the asymptotic states corresponding to the colliding system in the absence of the interaction V

$$|\Psi_i> \; = \; |\psi_{1s}> |K_i>, \tag{8}$$

$$|\Psi_f> \; = \; |\psi_{1s}> |K_f> . \tag{9}$$

Here $|\psi_{1s}>$ denotes the ground state of a hydrogen atom and $|K_{i,f}>$ are plane waves. The initial and final energies of the electron-atom system are

$$\mathcal{E}_i \; = \; \mathrm{E}_{1s} + \frac{p_i^2}{2}, \tag{10}$$

$$\mathcal{E}_f \; = \; \mathrm{E}_{1s} + \frac{p_f^2}{2}, \tag{11}$$

where E_{1s} is the unperturbed ground state energy and $p_{i(f)}$ is the initial (final) momentum of the projectile.

Our goal is to study two-photon processes leading to the same final energy of the scattered electron, given by equation (2). These processes are described by the following transition matrix element

$$\begin{aligned} T_{if}^{(\pm 2)} \; &= \; \frac{\mathcal{A}_0^2}{4} < \chi_f^- | \vec{\varepsilon} \cdot (\vec{p} + \vec{P}) \; G^+(\mathcal{E}_i \pm \omega) \; \vec{\varepsilon} \cdot (\vec{p} + \vec{P}) | \chi_i^+ > \\ &+ \; e^{\mp i\varphi} \, \frac{\mathcal{A}_0 \mathcal{A}_0'}{4} [< \chi_f^- | \vec{\varepsilon} \cdot (\vec{p} + \vec{P}) \; G^+(\mathcal{E}_i \pm 3\omega) \; \vec{\varepsilon}\,' \cdot (\vec{p} + \vec{P}) | \chi_i^+ > \\ &\qquad + < \chi_f^- | \vec{\varepsilon}\,' \cdot (\vec{p} + \vec{P}) \; G^+(\mathcal{E}_i \mp \omega) \; \vec{\varepsilon} \cdot (\vec{p} + \vec{P}) | \chi_i^+ >], \end{aligned} \tag{12}$$

which is related to the S-matrix element (4). The upper sign corresponds to the process in which the energy of the scattered electron is increased by 2ω and the lower sign to the case in which the energy is decreased by 2ω. For the sake of simplicity, we discuss here the significance of the two terms in Eq.(12) only for the case $E_f > E_i$, represented in Fig.1. The first line in equation (12), which is proportional to the intensity of the fundamental field, represents the transition matrix element corresponding to the process involving the absorption of two *identical* photons and we denote it by T_a. The other term is proportional to the potential vector of both components of the field (1) and it is connected to the diagrams

in Fig.1(b). It involves two *different* photons: the harmonic photon is absorbed and the fundamental one is emitted; we denote it by T_b. *Note that any high order correction to these leading terms are, at least, of the fourth order in the field.* A similar analysis may be done for $E_f < E_i$. In order to describe the process we are interested in, we match the individual matrix elements, T_a and T_b; the match involves the *relative phase* φ. One may write

$$T_{i,f}^{(\pm 2)} = T_a + e^{\mp i\varphi} T_b. \tag{13}$$

The evaluation of individual matrix elements was already extensively discussed in the literature; it is based on the 'two-potential' formalism used by Kracke *et al* (1994) for two identical photons and by Cionga and Buică (1998) for two different photons. In these works the authors restrict themselves to the domain of high scattering energies, therefore the first Born approximation is used to treat electron-atom scattering. In this way, the evaluation of the transition matrix element is made in the third order perturbation theory: the second order in the electric field and the first order in the scattering potential, V. When all the involved Feynman diagrams are included, every transition matrix element for a two-photon process may be written as the sum of three terms. For example, the process given in Fig.1(a) is described by

$$T_a = T_P^a + T_M^a + T_A^a \tag{14}$$

and a similar relation may be written for the process in Fig.1(b). $T_P^{a(b)}$, $T_M^{a(b)}$, and $T_A^{a(b)}$ account for the electronic, mixed, and atomic contributions, respectively. Each contribution is connected to specific Feynman diagrams as discussed by Kracke *et al* (1994) for identical photons and by Cionga and Buică (1998) for different photons. The angular structure of the these contributions, as well as their dependence on the frequencies and on the momentum transfer, are analyzed in the same papers. We mention that the analytic expression of T_a is the same for $\Delta E_\pm = E_f - E_i = \pm 2\omega$. However, for the two different signs of $\Delta E_\pm$ this expression is computed using different values of the parameter of the Green's function in Eq.(12). The same statements are true for T_b.

The differential cross section for the scattered electrons with the final energy (2) is then given by

$$\frac{d\sigma(\pm 2)}{d\Omega} = (2\pi)^4 \frac{p_f}{p_i} |T_{if}^{(\pm 2)}|^2, \tag{15}$$

being proportional to

$$|T_{if}^{(\pm 2)}|^2 = |T_a|^2 + |T_b|^2 + 2Re\left(T_a^* T_b e^{\mp i\varphi}\right). \tag{16}$$

The third term in the last expression is the interference one and it depends on the phase difference, φ. For bichromatic fields whose frequencies satisfy the relation $2\omega < |E_{1s}|$ both individual matrix elements, T_a and T_b, are real (Cionga and Buică 1998), therefore the phase dependence is simpler:

$$\frac{1}{I^2}\frac{d\sigma(\pm 2)}{d\Omega} = (2\pi)^4 \frac{p_f}{p_i}\left[\mathcal{T}_a^2 + \frac{I'}{I}\mathcal{T}_b^2 + 2\sqrt{\frac{I'}{I}}\mathcal{T}_a\mathcal{T}_b \cos\varphi\right]. \tag{17}$$

In this relation we have chosen to explicitly display the intensity dependence of the differential cross section, using the relations $T_a = I\ \mathcal{T}_a$ and $T_b = \sqrt{II'}\mathcal{T}_b$. I is the intensity of the laser field and I' that of the harmonic. Due to the power low, valid in the perturbative regime, we prefer to normalize our results to the square of the laser intensity. We note that in this case the differential cross section (17) is symmetric with respect to $\varphi = \pi$.

III. RESULTS AND DISCUSSION

In order to investigate the effects of the relative phase on laser assisted signals in free-free transitions, we carried out numerical calculations of the differential cross section (17) for fast projectiles, $E_i = 100$ eV, in the domain of optical frequencies. We illustrate our results for the frequency of Nd:YAG laser, $\omega = 1.17$ eV. Both components of the field (1) are linearly polarized, namely $\vec{\varepsilon} = \vec{\varepsilon}\,' \equiv \vec{p}_i/p_i$; $\vec{\varepsilon}$ defines the Oz axis. We have focused our attention on the study of phase effects at small scattering angles, where the dressing of the target is important and all three terms in Eq.(14) do contribute (Kracke *et al* 1994, Cionga and Buică 1998). The numerical evaluation of the individual transition matrix elements T_a and T_b is based on analytic expressions involving series of hypergeometric functions (Cionga and Florescu 1992, Cionga and Buică 1998).

Figures 2(a) and (b) are three dimension plots: the differential cross sections (17) are shown as a function of the scattering angle, θ, and of the relative phase, φ, for equal intensities, $I = I'$. *The panel (a) corresponds to* $\Delta E_+ = 2\omega$ *and the panel (b) to* $\Delta E_- =$

-2ω. In order to understand the "modulations" of these "surfaces", we note that the general structure of the differential cross section (17) is given by

$$\frac{1}{I^2}\frac{d\sigma(\pm 2)}{d\Omega} \sim \mathcal{L}(\theta) + \mathcal{L}'(\theta)\cos\varphi, \tag{18}$$

where

$$\begin{aligned} \mathcal{L}(\theta) &= \mathcal{T}_a^2 + \frac{I'}{I}\mathcal{T}_b^2, \\ \mathcal{L}'(\theta) &= 2\sqrt{\frac{I'}{I}}\mathcal{T}_a\mathcal{T}_b, \end{aligned} \tag{19}$$

with $\mathcal{L}(\theta) \geq 0$. As well as the individual matrix elements, T_a and T_b, these two quantities depend on the scattering angle. They also depend on the momentum transfer of the projectile and on the field frequencies, ω and 3ω. *The two deep minima present in Figs.2 (a) and (b) for the same relative phase,* $\varphi = 180^0$, *occur because* $\mathcal{L} \simeq \mathcal{L}'$ *for* $\theta \simeq 11^0$ *when* $\Delta E_+ = 2\omega$ *and for* $\theta \simeq 6^0$ *when* $\Delta E_- = -2\omega$.

More information is revealed in figure 3, which displays the differential cross section, $d\sigma(+2)/d\Omega/I^2$, as a function of the relative phase, φ, for six values of the scattering angle, belonging to the domain where the dressing is important. The parameters are the same as in Fig.2(a). For a given scattering angle, when the harmonic is out of phase with respect to the fundamental ($\varphi \neq 0$), the laser assisted signal is increased or decreased depending on the sign of $\mathcal{L}'$, as shown by Eq.(18). For all θ values for which one of the individual transition matrix elements, T_a or T_b, vanishes due to a dynamical interference between the three terms in Eq.(14), the laser assisted signal is φ-independent. This is the case for example at $\theta \simeq 13.4^0$, where $T_b = 0$. The first three changes of curvature between the panels (a) and (b), (b) and (c), and (c) and (d) are due to the three zeroes of the other matrix element, T_a; they are located at $\theta \simeq 4.4^0, 6.4^0$ and 10.25^0, respectively. More general, the φ-dependence change its curvature after every zero of the individual matrix elements that is due to cancellation between electronic, mixed, and atomic contributions (14). *There are only two changes of curvature in Fig.2(b) because only two such cancellations take place if* $\Delta E_- = -2\omega$, *one for* T_a *and the other one for* T_b.

The features discussed in the previous paragraph are the signature of the dressing of the target. When this dressing is neglected, the shape of the curve giving the φ-dependence

does not depend on the scattering angle, as one can see following the dotted curves in Fig.3. These curves represent $d\sigma(+2)/d\Omega/I^2$ computed in the approximation that only the first term is kept in Eq.(14) and its equivalent for T_b. We remind that this approximation is the generalization of Bunkin and Fedorov formula (1965) for a bichromatic field. Such calculations were carried out by Varró and Ehlotzky (1993a) in a different regime of frequencies and energies, therefore no specific comparison is made with that work. When the dressing is neglected, the θ-dependence factorizes out as follows:

$$\frac{1}{I^2}\frac{d\sigma(\pm 2)}{d\Omega} \simeq \left[f_{el}^{B1}(q)\right]^2 \frac{|(\vec{\varepsilon}\cdot\vec{q})|^4}{2^6\omega^8}\left[1+\frac{4}{81}\frac{I'}{I}-\frac{4}{9}\sqrt{\frac{I'}{I}}\cos\varphi\right], \tag{20}$$

where $\vec{q}$ is the momentum transfer of the projectile and $f_{el}^{B1} = 2\left(q^2+8\right)/\left(q^2+4\right)^2$ is the first Born approximation of the transition amplitude for elastic scattering on the potential V. We point out that, for $\theta = \arccos(p_i/p_f)$, the scalar product $\vec{\varepsilon}\cdot\vec{q}$ vanishes and therefore the differential cross section (20) vanishes, too. At large scattering angles, where the target dressing does not contribute significantly anymore, the full and the dotted curves become closer and closer, as one can see for θ=20^0.

An important parameter that has an influence on the quantum interference between the two paths in Fig.1 is the ratio between the intensities of the two field components, as one can see from Eq.(17). In Figure 3, when the dressing is neglected and $I = I'$, the laser assisted signal (dotted curves) is always increased if $\varphi \neq 0$. For other ratios I'/I the φ-dependence may be more complicated. In Figure 4 we have represented $d\sigma(+2)/d\Omega$, normalized with respect to I^2, as a function of the scattering angle, θ, for four different values of the relative phase: $\varphi = 0; \pi/4; \pi/2; \pi$. In each panel there are three curves, which correspond to the following intensity ratios: $I'/I = 1;\ 10^{-1};\ 10^{-2}$. When $\varphi = 0$ and this ratio gets smaller and smaller, the differential cross section becomes closer and closer to that given by the path (a) in Fig.1. In particular, for $I'/I = 10^{-2}$ the three zeroes of T_a discusses previously can be seen at the locations mentioned before. On the contrary, with the increasing of the harmonic intensity, one should recover the differential cross sections given by the path (b).

IV. CONCLUSION

In this paper we have studied the effect of the relative phase between the harmonic (3ω) and the fundamental field (ω) on two-photon free-free transitions in laser-assisted electron-hydrogen scattering. Using third order perturbation theory and taking into account all the involved Feynman diagrams, we have evaluated the differential cross sections for scattered electrons of energy $E_f = E_i \pm 2\omega$. The interference between the two quantum paths leading to the foregoing final energies was modulated by the relative phase. The signature of the target modification in the bichromatic field was discussed for fast projectiles and low field intensities in the domain of small scattering angles, where the dressing is important. We stress that whenever the target dressing can not be neglected, it influences significantly the phase effects.

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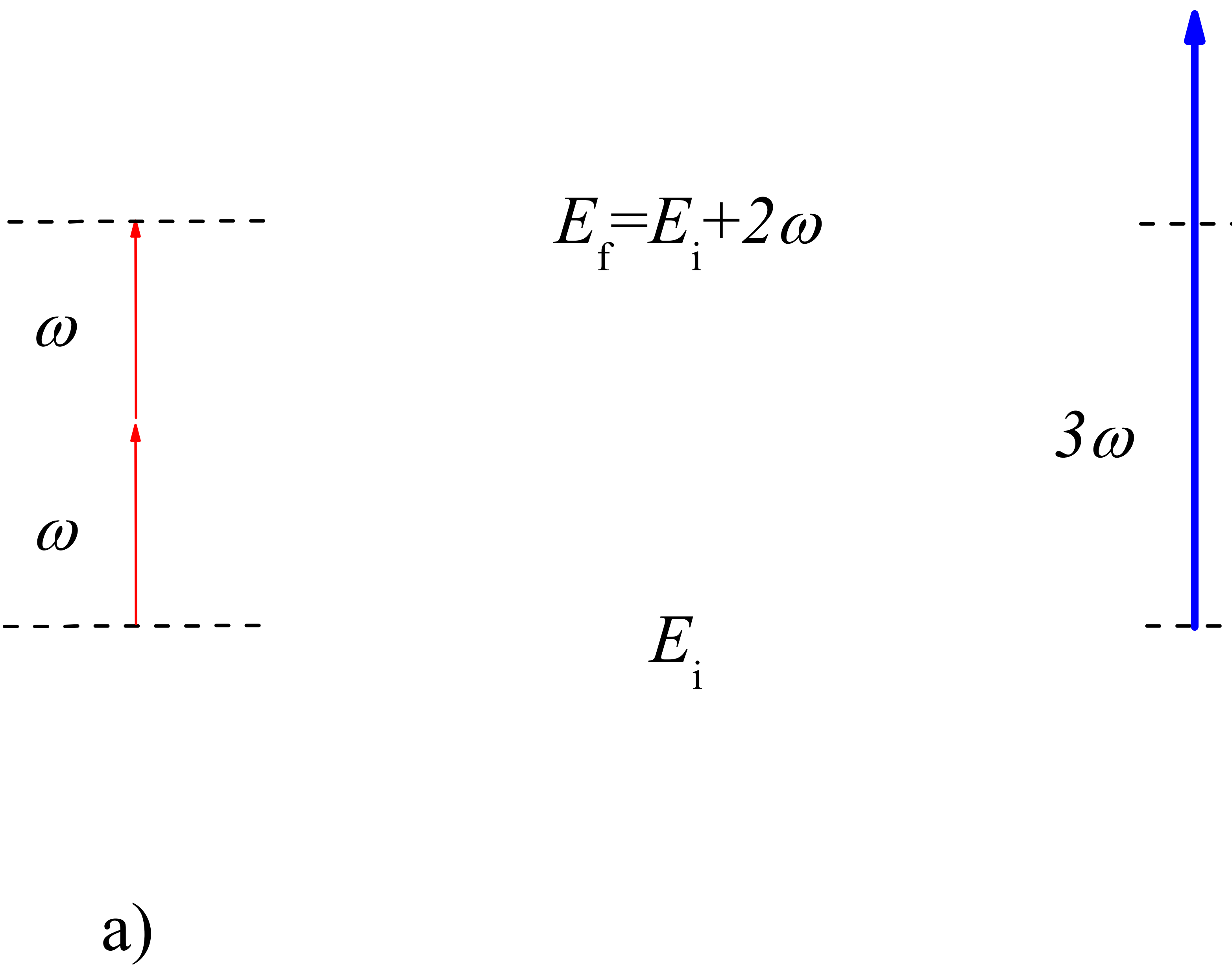
Figure Captions

- Fig.1: Energy diagrams schematically representing two-photon free-free transitions between the initial state, in which the projectile has the energy E_i, and the final one, in which it has the energy $E_f = E_i + 2\omega$. (a) corresponds to the absorption of two photons of frequency ω. (b) corresponds to the absorption of the harmonic and the emission of the laser photon. The laser photons are represented by thin lines, the harmonic photons by thick lines.

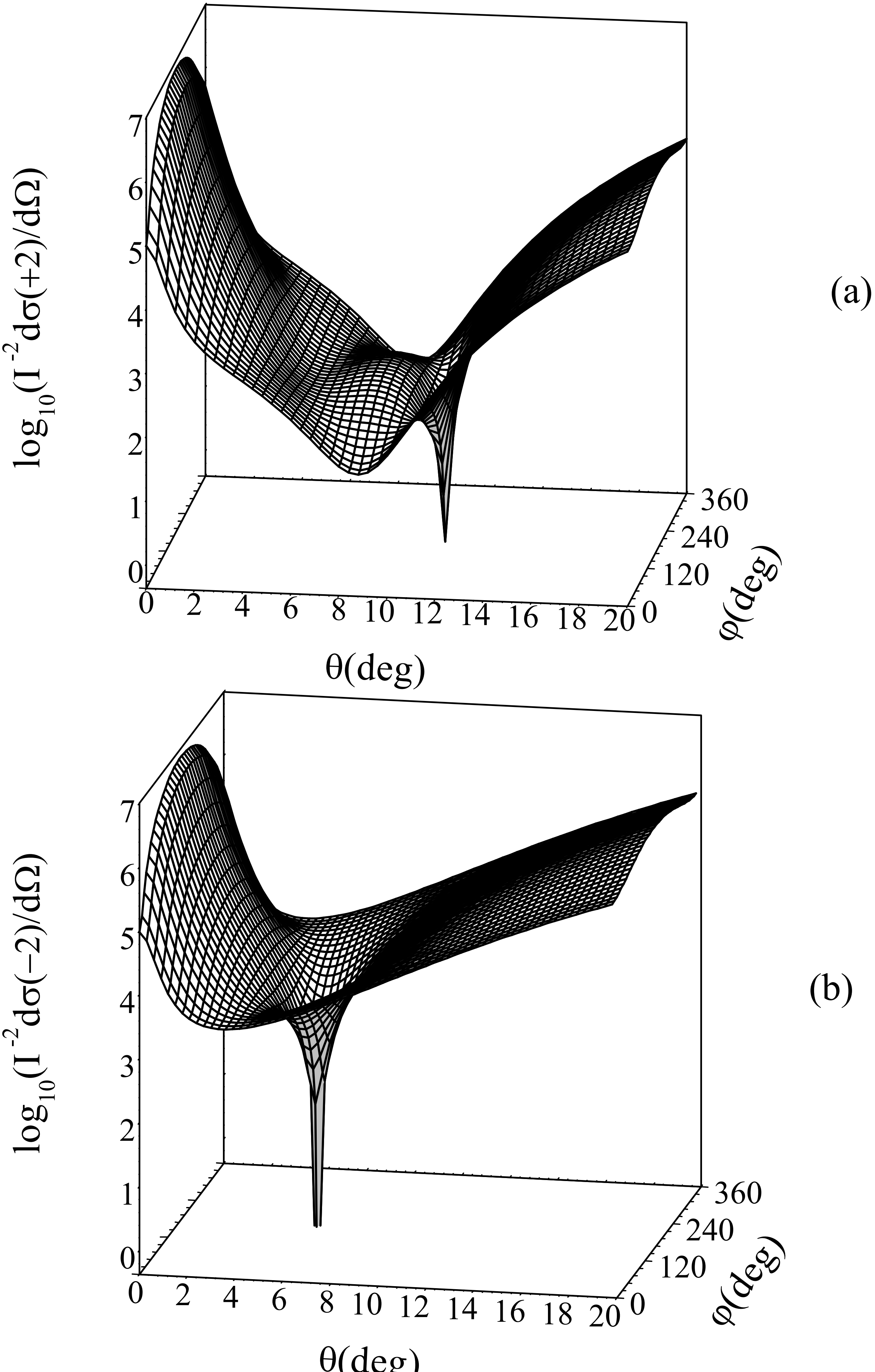
- Fig.2(a): $d\sigma(+2)/d\Omega/I^2$ in logarithmic scale, as a function of the scattering angle, θ, and the relative phase, φ. The initial energy is $E_i = 100$ eV and the laser frequency is $\omega = 1.17$ eV. The harmonic intensity is chosen equal to that of the laser. (b), idem fig. 2(a) but $d\sigma(-2)/d\Omega/I^2$.

- Fig.3: $d\sigma(+2)/d\Omega/I^2$ in logarithmic scale, as a function of the relative phase, φ, is represented by full lines for six values of the scattering angle. The dotted lines represent the same quantity when the target dressing is completely neglected. The parameters are the same as in Fig.2.

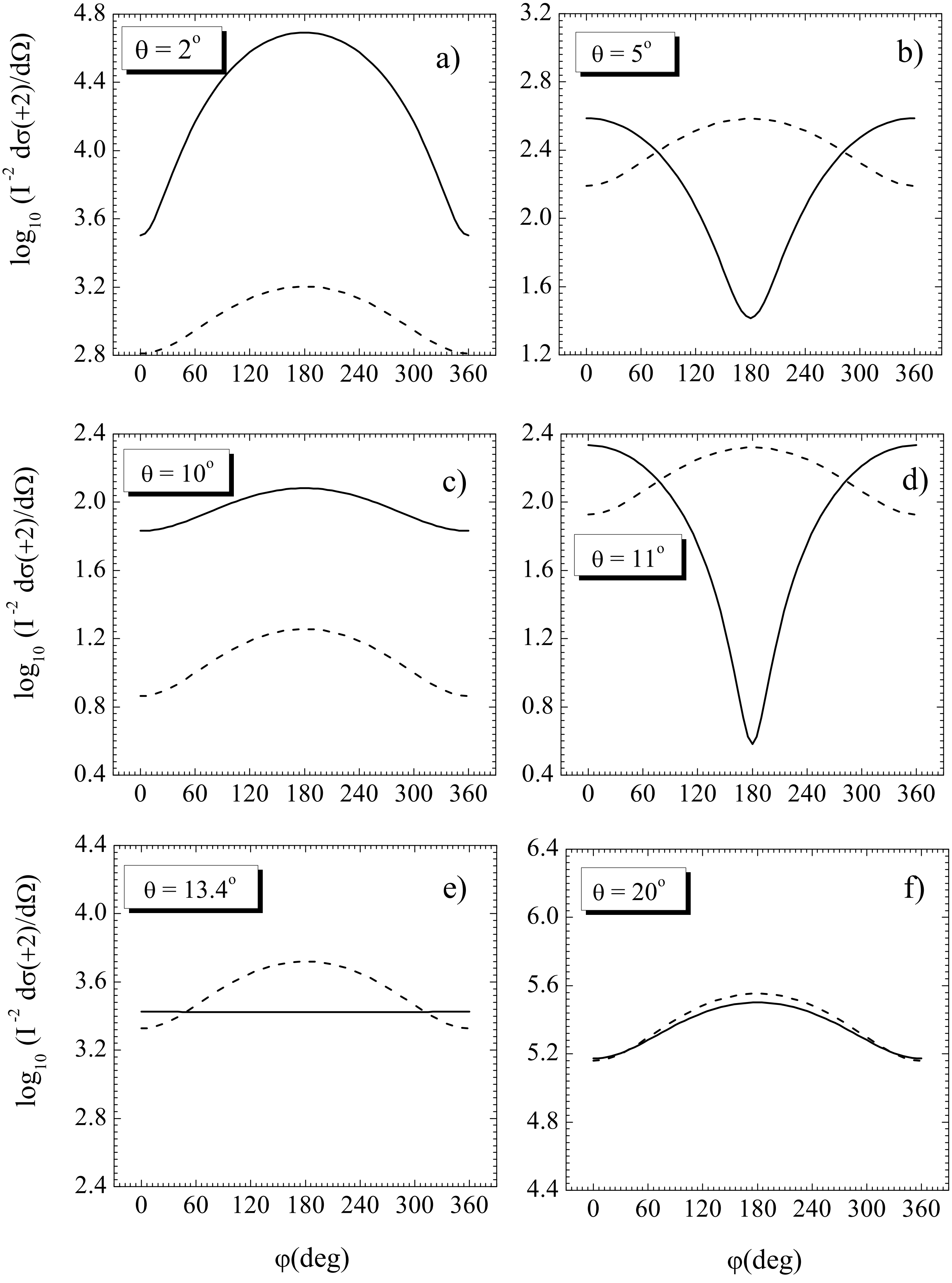
- Fig.4: $d\sigma(+2)/d\Omega/I^2$ in logarithmic scale, as a function of the scattering angle, θ, for four values of the relative phase: $\varphi = 0$, $\varphi = \pi/4$, $\varphi = \pi/2$, and $\varphi = \pi$. The initial energy is $E_i = 100$ eV, and the laser frequency is $\omega = 1.17$ eV. The intensity of the harmonic field represents 1% of the laser intensity (full lines), 10% (dotted-dashed lines), and 100% (dashed lines).



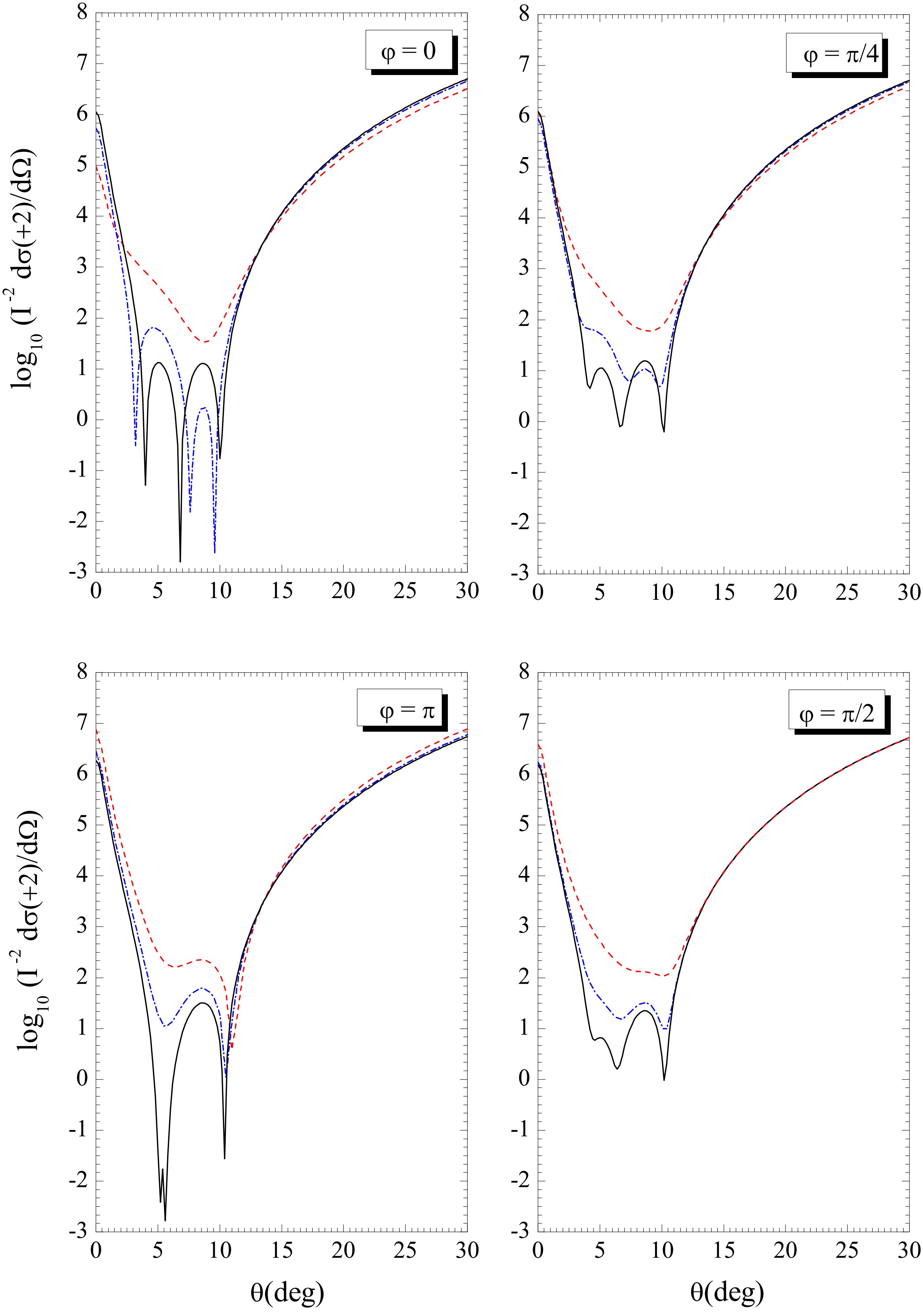
$E_f = E_i + 2\omega$
ω
ω
E_i
3ω
a)



log10(I^-2 dσ(+2)/dΩ)
0
1
2
3
4
5
6
7
0 2 4 6 8 10 12 14 16 18 20
θ(deg)
0
120
240
360
φ(deg)
(a)
log10(I^-2 dσ(−2)/dΩ)
0
1
2
3
4
5
6
7
0 2 4 6 8 10 12 14 16 18 20
θ(deg)
0
120
240
360
φ(deg)
(b)



θ = 2°
a)
θ = 5°
b)
θ = 10°
c)
θ = 11°
d)
θ = 13.4°
e)
θ = 20°
f)
log10 (I^-2 dσ(+2)/dΩ)
φ(deg)



φ = 0
φ = π/4
φ = π
φ = π/2
log₁₀ (I⁻² dσ(+2)/dΩ)
θ(deg)